Thickness dependent thermal capacitance of thin films with rough boundaries

Palasantzas, G

Published in:
Solid State Communications

DOI:
10.1016/S0038-1098(02)00184-9

PERGAMON

Solid State Communications 122 (2002) 523–526

solid state communications

www.elsevier.com/locate/ssc

Thickness dependent thermal capacitance of thin films with rough boundaries

George Palasantzas*

Department of Applied Physics, Materials Science Centre, University of Groningen, Nijenborgh 4, 9747 AG Groningen, The Netherlands

Received 15 February 2002; received in revised form 29 April 2002; accepted 29 April 2002 by J.H. Davies

Abstract

We investigated the thickness dependence of the thermal capacitance of thin films with evolving boundary roughness as a function of film thickness. Besides dynamic roughness evolution, also thickness variations of the film thermal conductivity were taken into account for the more general case of polycrystalline films. Nevertheless, the roughness evolution with film thickness is shown to be the dominant factor, modified by details of the corresponding scattering mechanisms that determine charge and heat carrier transport at low film thickness in comparison with the heat carrier mean bulk mean free path. © 2002 Elsevier Science Ltd. All rights reserved.

PACS: 65.40.+g; 67.80.Gb; 68.35.BS

Keywords: D. Heat capacity; D. Heat conduction; A. Surfaces and interfaces; A. Thin films

Although new device geometries require the growth of high quality thin films, kinetic effects during film growth can induce substantial roughening depending on the material, substrate, and deposition conditions. Deviations of surfaces/interfaces from flatness, as well as the presence of material defects (e.g. dislocations, impurities, etc.) may alter operation characteristics of microelectronic devices [1,2]. Under this framework surface/interface disorder effects on thermo-electrical properties of thin films are of potential importance in the field of microelectronics.

For example, the presence of a rough metal/insulator interface was shown to influence the field breakdown mechanism [3], as well as the capacitance and leakage currents in thin film capacitors [4]. Random rough surfaces influence the image potential of a charge situated in the vicinity of the vacuum/dielectric interface [5–8]. Such roughness effects could have a strong influence in inversion layers at a semiconductor/oxide interface since they may cause a shift of electronic levels [5–8] and consequently alter the device operation. In addition, surface/interface roughness has been shown to influence strongly the electrical conductivity of semiconducting and metallic thin film [9–15].

Besides electrical properties, thermal management problems in opto-electronic devices has been also a topic of intense research [16–19]. Indeed, the lifetime of metallic interconnects in integrated circuits depends strongly on the operating temperature because of resistive heating temperature increment. In photothermal analyses [20], metallic films deposited on dielectric substrates were utilised to deduce the dielectric thermal properties. The thermal conductivity $K_{\mathrm{Thc}}(h)$ of thin films is also less than its bulk value due to heat carrier surface and grain boundary scattering, which was confirmed also by experiment [16,20,21]. Finally, it has been shown that surface/interface roughness causes an increase of the film thermal capacitance $\langle C \rangle$ depending on specific roughness details [22].

Although roughness strongly affects the thermal capacitance as a function of film thickness, a precise determination of the actual effect requires detailed knowledge of the thickness dependence of the involved roughness parameters during film growth [22]. Besides roughness variation with film thickness, also variation of the film thermal conductivity $K_{\mathrm{Thc}}(h)$ with film thickness has to be taken into account since $\langle C \rangle \sim K_{\mathrm{Thc}}(h)$. This will be the topic under detailed investigation in the present work for the case of

* Tel.: +31-50363-4272-31; fax: +31-50-363-4881.
E-mail address: g.palasantzas@phys.rug.nl (G. Palasantzas).

PII: S0038-1098(02)00184-9

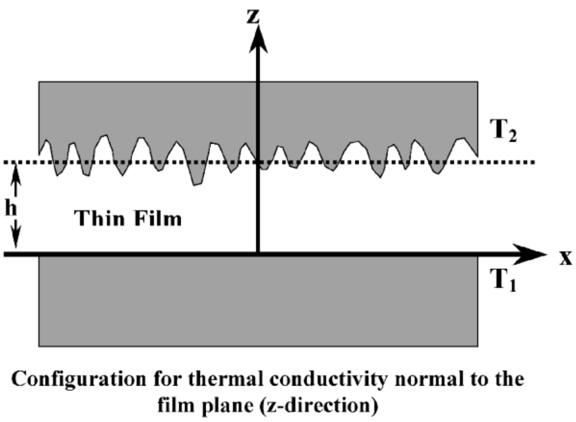

Fig. 1. Schematics of a thin film with a single rough boundary and average thickness h where thermal conduction normal to the film plane is assumed.

mound surface roughness observed in thin films grown under the influence of significant Schwoebel barrier [23–27].

We consider a film of thickness h with only one surface rough held at temperature T_2, and the other one smooth held at temperature $T_1 < T_2$ (Fig. 1) under steady heat flow conditions ($\partial T/\partial t = 0 \Leftrightarrow \nabla^2 T(x,y,z) = 0$) between the film boundaries. For the case of a random rough boundary with weak roughness ($|\nabla h| < 1$ and $w \ll h$), the film capacitance is given in Fourier space notation by [22]

$$\langle C\rangle = \tilde{C}_{\mathrm{B}}\frac{K_{\mathrm{Thc}}(h)}{K_{\mathrm{Bulk}}}M(h), \qquad \tilde{C}_{\mathrm{B}} = K_{\mathrm{Bulk}}\tau \tag{1}$$

$$M(h) = \frac{1}{h^2}\left\{1 + \frac{(2\pi)^4}{A}\left[2\int_0^{K_c} k^2\langle|h(k)|^2\rangle \mathrm{d}^2k + \frac{2\pi}{h}\int_0^{K_c}\frac{\cosh(kh)}{\sinh(kh)}k\langle|h(k)|^2\rangle \mathrm{d}^2k\right]\right\} \tag{2}$$

with $K_c = \pi/a_0$, where a_0 is a lower roughness cut-off of the order of the atomic spacing, τ the time that the heat flux is passing through the film, $\langle|h(k)|^2\rangle$ the roughness spectrum, and K_{Bulk} the bulk thermal conductivity. $K_{\mathrm{Thc}}(h)$ is the (thickness dependent) thermal conductivity normal to the film plane. Note that in the derivation of Eqs. (1) and (2) we have assumed translation invariant surfaces ($\langle h(k)h(k')\rangle = [(2\pi)^4/A]\langle|h(k)|^2\rangle\delta(k+k')$) where A denotes the average flat macroscopic surface area. Moreover, the thermal conductivity $K_{\mathrm{Thc}}(h)$ in Eq. (1) is given for polycrystalline thin films by

$$K_{\mathrm{Thc}}(h) = K_{\mathrm{Bulk}}\left[1 + \frac{3}{2}\frac{L}{D}\frac{R}{1-R}\right]^{-1} \tag{3}$$

with L the electron mean free path (i.e. $L \sim 41$ nm for example Au [16]), R the electron reflection coefficient due to grain boundary scattering, and D the average grain size. The derivation of Eq. (3) is based on the form of the electrical conductivity normal to the film plane $\sigma_{\mathrm{Th}}(h) = \sigma_{\mathrm{Bulk}}[1 + \{3LR/2D(1-R)\}]^{-1}$ for isotropic polycrystalline bulk material with randomly orientated grains [28], and the use of the Wiedemann–Franz law assuming that electrons serve both as electrical and thermal carriers with phonons having negligible effect for metals [16]. This law was discovered initially for bulk materials, and has been proven to be valid also for thin films [17–19].

Furthermore, for the thermal capacitance calculations we will consider the case of mound roughness, which has been described in the past by the interface width w, the system correlation length ζ which determines how randomly the mounds are distributed on the surface, and the average mound separation λ [23–26]. Such a rough morphology can be described in Fourier space by the roughness spectrum $\langle|h(k)|^2\rangle$ [27]

$$\langle|h(k)|^2\rangle = \frac{A}{(2\pi)^5}\frac{w^2\zeta^2}{2}\mathrm{e}^{-(4\pi^2+k^2\lambda^2)(\zeta^2/4\lambda^2)}I_0(\pi k\zeta^2/\lambda) \tag{4}$$

with $I_0(x)$ the modified Bessel function of first kind and zero order. If $\zeta \geq \lambda$ the surface is characteristic to that caused by Schwoebel barrier effects [23–27]. Note that the height correlation function $C_{\mathrm{cor}}(r) \propto \int\langle|h(k)|^2\rangle \mathrm{e}^{-jkr}\mathrm{d}^2r$ for mound roughness has an oscillatory behaviour for $\zeta \geq \lambda$ (strong Schowebel barrier effect during film growth) leading to a characteristic satellite ring at $k = 2\pi/\lambda$ of the power spectrum $\langle|h(k)|^2\rangle$ [27].

For simplicity the thickness evolution of the roughness parameters (w, ζ, λ) was considered in terms of the relations $w = 0.5(h/10)^{0.24}$ nm, $\zeta = 10(h/10)^{0.26}$ nm, and $\lambda = 10(\mathrm{F})(\mathrm{h}/10)^{0.26}$ nm with F a parameter in order to distinguish for the cases $\lambda > \zeta$ and $\lambda < \zeta$. The choice of these thickness variations are suggested by experimental facts. For example, the average mound separation has been shown to evolve with film thickness (or growth time for fixed deposition rate) as $\lambda \propto h^{\delta}$ ($0.16 \leq \lambda \leq 0.26$) [23–26], and the rms roughness amplitude as $w \propto h^{\beta}$ ($\beta < 1$) [27]. Such a choice satisfies also the requirement of weak roughness ($|\nabla h| < 1$) or alternatively small rms local surface slope $\rho_{\mathrm{rms}} = \langle|\nabla h|^2\rangle^{1/2} < 1$ and small rms roughness amplitude $w \ll h$.

Fig. 2 shows the dependence of the local surface slope $\rho_{\mathrm{rms}} = \langle|\nabla h|^2\rangle^{1/2}$ on film thickness for various values of the parameter F. The oscillatory behaviour for $F = 0.5$ and thus $\lambda < \zeta$ is typical for mound roughness due to the characteristic satellite ring at $k = 2\pi/\lambda$ of the power spectrum $\langle|h(k)|^2\rangle$, that leads also to oscillations also of the corresponding real space correlation function. For $F \geq 1$ a behaviour characteristic to that of a typical Gaussian random roughness evolves with a continues decreasing local slope since w/λ and w/ζ decrease with increasing film thickness (w/λ and $w/\zeta \propto h^{-0.02} \to 0$ if $h \gg 1$).

Since from Eq. (5) we have $\langle|h(k)|^2\rangle \propto w^2$, the thermal capacitance $\langle C\rangle$ will have a simple dependence on the roughness amplitude w, while any complex dependence will arise as a function of the lateral length scales ζ and λ through the factor $M(h)$. Moreover, during the calculations of $\langle C\rangle$ we

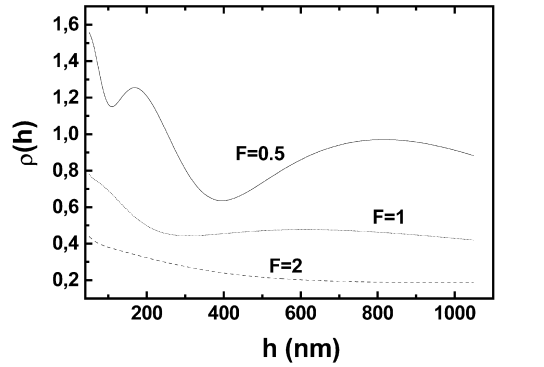

Fig. 2. Rms local surface slope $\rho_{\mathrm{rms}} = \langle |\nabla h|^2 \rangle^{1/2}$ as a function of the film thickness for $a_0 = 0.3$ nm, and various values of the coefficient F that controls the relative magnitude of the lateral length scales ζ and λ.

considered the average grain size D in Eq. (3) to be related to the lateral roughness parameters ζ and λ by means of the equation $2/D^2 = 1/\zeta^2 + 1/\lambda^2$ (accounting also for grain size changes with film thickness h).

Fig. 3 shows the dependence of the thermal capacitance ratio $\langle C \rangle / C_{\mathrm{B}}$ (with $C_{\mathrm{B}} = \tilde{C}_{\mathrm{B}} \times 10^4$) as a function of the film thickness for various values of the coefficient F or equivalently different magnitudes of ζ and λ. Indeed, for $\zeta > \lambda$ ($F < 1$) the thermal capacitance $\langle C \rangle$ decays much faster with increasing film thickness h while a weak oscillation is visible for low film thickness ($h < L/2$) as the arrow indicates which is a reminiscent from the local slope oscillatory behaviour in Fig. 2. For $\zeta \le \lambda$ ($F > 1$) the relative reduction of $\langle C \rangle$ is slower as the thickness increases especially for small system correlation lengths $\zeta \ll \lambda$ ($F \gg 1$).

In order to understand the influence of the thickness dependent terms $K_{\mathrm{Thc}}(h)$ and $M(h)$ on the thermal capacitance $\langle C \rangle$ we plot them in Fig. 4 separately as a function of film thickness. Indeed, with increasing thickness h, the roughness term $M(h)$ clearly decreases with roughness following closely the behaviour of the local surface slope

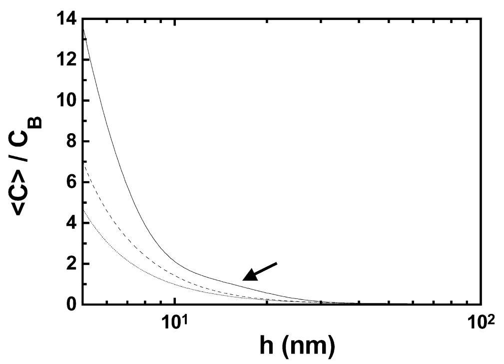

Fig. 3. Thermal capacitance ratio $\langle C \rangle / C_{\mathrm{B}}$ as a function of the film thickness h for various values of F, grain boundary reflection coefficient $R = 0.1$, $L = 41$ nm, and $a_0 = 0.3$ nm. Solid line $F = 0.5$, dashes $F = 1$, dots $F = 2$.

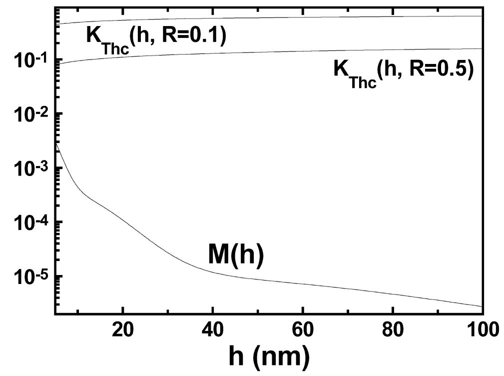

Fig. 4. Calculation of $M(h)$ and $K_{\mathrm{Thc}}(h)$ as a function of film thickness h for $F = 0.5$, $L = 41$ nm, and $a_0 = 0.3$ nm. The other parameters are indicated.

$\rho_{\mathrm{rms}} = \langle |\nabla h|^2 \rangle^{1/2}$, while the thermal capacitance $K_{\mathrm{Thc}}(h)$ increases approaching values for large thickness close to the bulk thermal conductivity. Since the change of the roughness term $M(h)$ is much stronger than that of $K_{\mathrm{Thc}}(h)$, not only in magnitude but also in rate of decrement, the roughness contribution will be the dominant one as a function of film thickness. Similar is the behaviour for various mean free paths L where however, $K_{\mathrm{Thc}}(h)$ increases with decreasing L.

Notably, with increasing reflection coefficient R (stronger electron scattering at grain boundaries) the thermal capacitance $\langle C \rangle$ decreases strongly in magnitude and at a fast rate for relatively small film thickness ($<L/2$; Fig. 5). On the other hand, as the inset of Fig. 5 indicates, with decreasing electron mean free path L the thermal capacitance $\langle C \rangle$ increases strongly in magnitude and at a faster rate with decreasing h. In comparison with the effect of R, the influence of the mean free path L is more drastic in the magnitude of $\langle C \rangle$ as Fig. 5 indicates.

Since in the expression of $M(h)$ the third term has low significance due to its $1/h$ dependence, neglecting this term

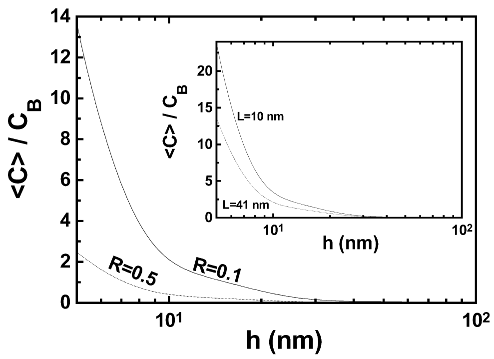

Fig. 5. Thermal capacitance ratio $\langle C \rangle / C_{\mathrm{B}}$ as a function of the film thickness h for $F = 0.5$, various grain boundary reflection coefficients R, $L = 41$ nm, and $a_0 = 0.3$ nm. The inset shows $\langle C \rangle / C_{\mathrm{B}}$ as a function of the film thickness h for $F = 0.5$, various mean free paths L, $R = 0.1$, and $a_0 = 0.3$ nm.

and extending the integral of the second term to infinity we obtain $M(h) \approx h^{-2}\{1 + w^2(\zeta^{-2} + \pi^2\lambda^{-2})\}$ which yields for the termal capacitance $\langle C \rangle$ the simple analytic expression

$$\frac{\langle C \rangle}{\tilde{C}_{\mathrm{B}}} \approx \frac{1}{h^2}\left[1 + w^2\left(\frac{1}{\zeta^2} + \frac{\pi^2}{\lambda^2}\right)\right]\left[1 + \frac{3}{2}\frac{L}{D(h)}\frac{R}{1-R}\right]^{-1} \tag{5}$$

Besides the case of mound roughness, similar results will be obtained for the case of self-affine roughness which is characterised by the roughness exponent H $(0 < H < 1)$, the rms roughness amplitude w, and the in-plane correlation length ξ [29–31]. Also in this case w and ξ evolve with film thickness as power-laws such that $w \propto h^b$ and $\xi \propto h^{1/z}$ (with $b < 1$ and $1/z < 1$) [29–31]. Qualitatively the thickness dependence of $\langle C \rangle$ will be similar to the case of mound roughness (which corresponds to $H \approx 1$) with Gaussian character $(\zeta < \lambda)$. Notably, if we consider in this case for $\langle |h(k)|^2 \rangle$ the Lorentzian model [32,33[1]] $\langle |h(k)|^2 \rangle = [A/(2\pi)^5][w^2\xi^2/(1 + ak^2\xi^2)^{1+H}]$ with $a = (1/2H)\times[1 - (1 + aK_c{}^2\xi^2)^{-H}]$, we obtain for $\langle C \rangle$ the simple expression (ignoring in Eq. (3) the third term due to its $1/h$ dependence)

$$\frac{\langle C \rangle}{\tilde{C}_{\mathrm{B}}} \approx \frac{1}{h^2}\left[1 + \frac{w^2}{a^2\xi^2}\left\{\frac{1}{1-H}\left[(1 + \mathrm{aKc}^2\xi^2)^{1-H} - 1\right] - 2a\right\}\right]\left[1 + \frac{3}{2}\frac{L}{D(h)}\frac{R}{1-R}\right]^{-1}. \tag{6}$$

In summary, we investigated the thickness dependence of the thermal capacitance of thin films with one smooth boundary and the other rather rough at nanometer length scales. Besides dynamic roughness evolution with film thickness, also thickness variation of the film thermal conductivity were taken into account for the case of polycrystalline films. The surface roughness contribution is the dominant one leading to a reduction of the thermal capacitance with increasing film thickness which is modified, however, by details of the corresponding scattering mechanisms that determine heat carrier transport at relatively low film thickness $(h < L)$.

Acknowledgments

I would like to acknowledge support from the 'Nederlandse Organisatie voor Wetenschappelijk Onderzoek (NWO)', and useful discussions with Dr G. Backx.

[1] Besides the simplicity of $\langle |h(k)|^2 \rangle$ it yields the analytic correlation function $C_{\mathrm{cor}}(r) = [w^2/a\Gamma(1+H)]\times(r/2a^{1/2}\xi)^H K_H(r/2a^{1/2}\xi)$ with $K_H(x)$ the second kind Bessel function of order H and $\Gamma(x)$ the Gamma function.

References

[1] S.M. Sze, Semiconductor Devices: Physics and Technology, Wiley, New York, 1985.
[2] K.A. McKinley, N.P. Sandler, Thin Solid Films 290 (1996) 440.
[3] J.H. Oh, Y.H. Lee, B.K. Ju, D.K. Shin, C.Y. Park, M.H. Oh, J. Appl. Phys. 82 (1997) 6203.
[4] Z.P. Zhao, G.-C. Wang, T.-M. Lu, G. Palasantzas, J.Th.M. de Hosson, Phys. Rev. B 60 (1999) 9127.
[5] T.S. Rahman, A.A. Maradudin, Phys. Rev. B 21 (1980) 504.
[6] G. Palasantzas, J. Appl. Phys. 82 (1997) 351.
[7] F.J. Ohkawa, Y. Uemura, Prog. Theor. Phys. Suppl. 57 (1975) 164.
[8] T. Ando, J. Phys. Soc. Jpn 39 (1975) 411.
[9] T. Ando, A.B. Fowler, F. Stern, Rev. Mod. Phys. 54 (1982) 437.
[10] G. Fishman, D. Calecki, Phys. Rev. Lett. 62 (1989) 1302.
[11] G. Fishman, D. Calecki, Phys. Rev. B 43 (1991) 11581.
[12] H. Sakaki, T. Noda, K. Hirakawa, K. Tanaka, T. Matsusue, Appl. Phys. Lett. 51 (1987) 1934.
[13] G. Palasantzas, J. Barnas, Phys. Rev. B 56 (1997) 7726.
[14] G. Palasantzas, J. Barnas, Phys. Status Solidi B 209 (1998) 319.
[15] J. Barnas, G. Palasantzas, J. Appl. Phys. 82 (1997) 3950.
[16] G. Chen, P. Hui, Appl. Phys. Lett. 74 (1999) 2942.
[17] J.C. Lambropoulos, S.D. Jacobs, S.J. Burns, L. Shaw-Klein, S.-S. Hwang, Thin Film Heat Transfer: Properties and Processing, vol. 184, ASME, New York, 1991, p. 21.
[18] T. Yamane, Y. Moi, S. Katayama, M. Todoki, J. Appl. Phys. 82 (1997) 1153.
[19] T. Startz, U. Schmidt, F. Volklein, Sens. Mater. 7 (1995) 395.
[20] Z.L. Wu, P.K. Kuo, L. Wei, S.L. Gu, R.L. Thomas, Thin Solid Films 236 (1993) 191.
[21] T.Q. Qui, C.L. Tien, Trans. ASME 115 (1993) 842.
[22] G. Palasantzas, J.Th.M. De Hosson, J. Appl. Phys. 89 (2001) 6130.
[23] M.D. Johnson, C. Orme, A.W. Hunt, D. Graff, J. Sudijono, L.M. Sander, B.G. Orr, Phys. Rev. Lett. 72 (1994) 116.
[24] M. Siegert, M. Plischke, Phys. Rev. Lett. 73 (1994) 1517.
[25] J.-K. Zuo, J.F. Wendelken, Phys. Rev. Lett. 78 (1997) 2791.
[26] J.A. Stroscio, D.T. Pierce, M.D. Stiles, A. Zangwill, Phys. Rev. Lett. 75 (1995) 4246.
[27] Y.-P. Zhao, H.-Y. Yang, G.C. Wang, T.-M. Lu, Phys. Rev. B 57 (1998) 1922.
[28] A. Knabchen, J. Phys. Cond. Mattter 3 (1991) 6989 See Eq. (18a) in this work which considers the case of normal to the film plane charge conduction.
[29] P. Meakin, Phys. Rep. 235 (1994) 1991.
[30] J. Krim, G. Palasantzas, Int. J. Mod. Phys. B 9 (1995) 599.
[31] F. Family, T. Viscek, Dynamics of Fractal Surfaces, World Scientific, Singapore, 1991.
[32] G. Palasantzas, Phys. Rev. B 48 (1993) 14472.
[33] G. Palasantzas, Phys. Rev. B 49 (1994) 5785 (E).